

Ac-cotunneling through an interacting quantum dot in a magnetic field

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We analyze inelastic cotunneling through an interacting quantum dot subject to an ambient magnetic field in the weak tunneling regime under a non-adiabatic time-dependent bias-voltage. Our results clearly exhibit photon-assisted satellites and an overall suppression of differential conductance with increasing driving amplitude, which is consistent with experiments. We also predict a zero-anomaly in differential conductance under an appropriate driving frequency.

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Recently, cotunneling¹ through discrete levels, i.e., a quantum dot (QD), has attracted much attention since it determines the intrinsic limitation of accuracy of single-electron transistors due to leakage, and since it also involves correlation effects, such as the Kondo effect.² It has also been reported experimentally³ and theoretically^{4,5,6,7} that external microwave irradiation can induce the occurrence of Kondo satellites and an overall suppression of the Kondo peak. However, there are few studies so far concerning time-dependent second-order cotunneling in the weak tunneling regime at temperatures above the Kondo temperature. About ten years ago, Flensberg presented an analysis for coherent photon-assisted cotunneling in a double-junction Coulomb blockade device in the adiabatic limit.⁸ In this letter, we will further study the cotunneling in an interacting QD when an ac bias-voltage is applied between two electrodes in the non-adiabatic regime.

We employ the s-d exchange Hamiltonian to model inelastic cotunneling through a QD in an ambient magnetic field, B , in the weak-coupling regime:

$$\begin{aligned} H &= H_0 + H_I, \\ H_0 &= \sum_{\eta\mathbf{k}\sigma} \varepsilon_{\eta\mathbf{k}}(t) c_{\eta\mathbf{k}\sigma}^\dagger c_{\eta\mathbf{k}\sigma} - \Delta_0 S^z, \\ H_I &= \sum_{\eta,\eta',\mathbf{k},\mathbf{k}'} J [(c_{\eta\mathbf{k}\uparrow}^\dagger c_{\eta'\mathbf{k}'\uparrow} - c_{\eta\mathbf{k}\downarrow}^\dagger c_{\eta'\mathbf{k}'\downarrow}) S^z \\ &\quad + c_{\eta\mathbf{k}\uparrow}^\dagger c_{\eta'\mathbf{k}'\downarrow} S^- + c_{\eta\mathbf{k}\downarrow}^\dagger c_{\eta'\mathbf{k}'\uparrow} S^+] + H_{\text{dir}}, \\ H_{\text{dir}} &= J_d \sum_{\sigma} (c_{L\mathbf{k}\sigma}^\dagger + c_{R\mathbf{k}\sigma}^\dagger) (c_{L\mathbf{k}\sigma} + c_{R\mathbf{k}\sigma}), \end{aligned} \quad (1)$$

where $c_{\eta\mathbf{k}\sigma}^\dagger$ ($c_{\eta\mathbf{k}\sigma}$) is the creation (annihilation) operator for electrons with momentum \mathbf{k} , spin- σ in lead η ($= L, R$). The energies $\varepsilon_{\eta\mathbf{k}}(t) = \varepsilon_{\eta\mathbf{k}}^0 + eV_\eta(t)$ include a rigid shift of the Fermi energy of the electrons in the leads due to the applied time-dependent bias-voltage $V_\eta(t) = V_\eta^0 + v_\eta \cos(\Omega t)$ with V_η^0 (v_η) being the amplitude of the dc(ac) part of the bias-voltage. Here, we assume that the Fermi energies of two leads are zero at equilibrium and $V_L^0 = -V_R^0 = eV_0/2$. $\Delta_0 = g_e \mu_B B$

is the static magnetic-field B -induced Zeeman energy. $\mathbf{S} \equiv (S^x, S^y, S^z)$ are Pauli spin operators of electrons in the QD [$S^\pm \equiv S^x \pm iS^y$], and J is the exchange coupling constant. H_{dir} is the potential scattering term with $2J_d = J$. As in our previous paper,⁹ we can rewrite the tunneling term, H_I in Eq. (1), as a sum of three products of two variables:

$$H_I = Q^z S^z + Q^+ S^- + Q^- S^+, \quad (2)$$

with the generalized coordinates $Q^{z(\pm)}$ of reservoir variables as

$$Q^z = \sum_{\eta,\eta'} Q_{\eta\eta'}^z = \sum_{\eta,\eta',\mathbf{k},\mathbf{k}'} J (c_{\eta\mathbf{k}\uparrow}^\dagger c_{\eta'\mathbf{k}'\uparrow} - c_{\eta\mathbf{k}\downarrow}^\dagger c_{\eta'\mathbf{k}'\downarrow}), \quad (3)$$

$$Q^+ = \sum_{\eta,\eta'} Q_{\eta\eta'}^+ = \sum_{\eta,\eta',\mathbf{k},\mathbf{k}'} J c_{\eta\mathbf{k}\uparrow}^\dagger c_{\eta'\mathbf{k}'\downarrow}, \quad (4)$$

$$Q^- = \sum_{\eta,\eta'} Q_{\eta\eta'}^- = \sum_{\eta,\eta',\mathbf{k},\mathbf{k}'} J c_{\eta\mathbf{k}\downarrow}^\dagger c_{\eta'\mathbf{k}'\uparrow}, \quad (5)$$

and $Q^{\hat{1}} = H_{\text{dir}}$. In the following, we will use units where $\hbar = k_B = e = 1$.

As in our previous studies of inelastic cotunneling through an interacting QD in the weak tunneling limit, we employ a generic quantum Langevin equation approach^{10,11,12} to establish a set of quantum Bloch equations for the description of the dynamics of a single spin [modeled by Eq. (1)] explicitly in terms of the response and correlation functions of free reservoir variables. This procedure provides explicit analytical expressions for the nonequilibrium magnetization and cotunneling current for arbitrary dc bias-voltage and temperature.^{9,13} Here, we generalize our previous derivations to the time-dependent case in the non-adiabatic and high-frequency regime.

In the derivation, we proceed with the Heisenberg equation of motion for the Pauli spin operators and the lead operators, and then formally integrate these equations from the initial time 0 to t exactly to all orders of J . Next, under the assumption that the time scale of

decay processes is much slower than that of free evolution, we replace the time-dependent operators involved in the integrals of these EOM's approximately in terms of their free evolutions. Thirdly, these EOM's are expanded in powers of J up to second order, resulting in non-Markovian dynamic equations for the time evolution of the QD spin variables in a compact form:

$$\begin{aligned} \dot{S}^z(t) = & -2 \int_{-\infty}^t dt' (e^{-i\Delta_0\tau} + e^{i\Delta_0\tau}) C(t, t') S^z(t') \\ & - \int_{-\infty}^t dt' (e^{-i\Delta_0\tau} - e^{i\Delta_0\tau}) R(t, t'), \end{aligned} \quad (6)$$

$$\begin{aligned} \dot{S}^\pm(t) = & \mp i\Delta_0 S^\pm(t) - 2 \int_{-\infty}^t dt' C(t, t') S^\pm(t') \\ & - 2 \int_{-\infty}^t dt' e^{\mp i\Delta_0\tau} C(t, t') S^\pm(t'), \end{aligned} \quad (7)$$

with $\tau = t - t'$. The correlation function, $C(t, t')$, and the response function, $R(t, t')$, of free reservoir variables (tagged by subscript "o") are defined as:

$$C(R)(t, t') = \frac{1}{2}\theta(\tau)\langle[Q_o^\pm(t), Q_o^\mp(t')]\rangle_{+(-)}. \quad (8)$$

Special attention must be paid to the free reservoir variables due to the time-dependent energies $\varepsilon_{\eta\mathbf{k}}(t)$:

$$c_{\eta\mathbf{k}\sigma}^o(t) = e^{-i\int_{t'}^t \varepsilon_{\eta\mathbf{k}}(\tau)d\tau} c_{\eta\mathbf{k}\sigma}(t'). \quad (9)$$

Therefore, the kernels $C(R)(t, t')$ become *double-time-dependent* functions due to the lack of time-translation-invariance stemming from the ac-bias:

$$\begin{aligned} C(R)(t, t') = & \frac{1}{2}\theta(\tau)\langle[Q_o^+(t), Q_o^-(t')]\rangle_{\pm} \\ = & \frac{1}{2}\theta(\tau)J^2 \sum_{\eta, \eta', \xi, \xi'} \sum_{\mathbf{k}, \mathbf{k}', \mathbf{q}, \mathbf{q}'} \langle[c_{\eta\mathbf{k}\uparrow}^\dagger(t)c_{\eta'\mathbf{k}'\downarrow}(t), \\ & c_{\xi\mathbf{q}\downarrow}^\dagger(t')c_{\xi'\mathbf{q}'\uparrow}(t')]\rangle_{\pm} \\ = & \frac{1}{2}\theta(\tau)J^2 \\ & \times \sum_{\eta, \eta', \xi, \xi'} \sum_{\mathbf{k}, \mathbf{k}', \mathbf{q}, \mathbf{q}'} e^{i\int_{t'}^t d\tau [\varepsilon_{\xi'\mathbf{q}'}(\tau) - \varepsilon_{\xi\mathbf{q}}(\tau)]} \\ & \times \left[\langle c_{\eta\mathbf{k}\uparrow}^\dagger(t)c_{\xi'\mathbf{q}'\uparrow}(t) \rangle \langle c_{\eta'\mathbf{k}'\downarrow}(t)c_{\xi\mathbf{q}\downarrow}^\dagger(t) \rangle \right. \\ & \left. \pm \langle c_{\xi\mathbf{q}\downarrow}^\dagger(t)c_{\eta'\mathbf{k}'\downarrow}(t) \rangle \langle c_{\xi'\mathbf{q}'\uparrow}(t)c_{\eta\mathbf{k}\uparrow}^\dagger(t) \rangle \right] \\ = & \frac{1}{2}\theta(\tau) \sum_{\eta} g \int d\epsilon d\epsilon' e^{i(\epsilon - \epsilon')\tau} \\ & \times \{f_{\eta}(\epsilon)[1 - f_{\eta}(\epsilon')] \pm f_{\eta}(\epsilon')[1 - f_{\eta}(\epsilon)]\} \\ & + \frac{1}{2}\theta(\tau)g \int d\epsilon d\epsilon' \left[e^{i(\epsilon - \epsilon')\tau} \right. \\ & \times e^{i\int_{t'}^t d\tau' V_{ac} \cos(\Omega\tau')} \pm e^{-i(\epsilon - \epsilon')\tau} \\ & \times e^{-i\int_{t'}^t d\tau' V_{ac} \cos(\Omega\tau')} \left. \right] \{f_L(\epsilon)[1 - f_R(\epsilon')] \\ & \pm f_R(\epsilon')[1 - f_L(\epsilon)]\}, \end{aligned} \quad (10)$$

with $g \equiv J^2 \rho_0^2$, $V_{ac} = v_L - v_R$, and the Fermi-distribution function is $f_{\eta}(\epsilon) = [1 + e^{(\epsilon - \mu_{\eta})/T}]^{-1}$ (T is the temperature). Here we assume the two electrodes to be Markov-type reservoirs with a constant density of states ρ_0 . The

kernels reduce exactly to our previous results, Eq. (B8) in Ref. 9, if there is no ac-bias or with the same ac amplitude in the left and right leads.

In the presence of a periodic ac-bias, the spin variables $S^{z(\pm)}(t)$ naturally depend periodically on t with a period $\mathcal{T}_{ac} = 2\pi/\Omega$. As a result, the full solutions of Eqs. (6) and (7) can be formally written as a superposition of all harmonics

$$S^{z(\pm)}(t) = \sum_{n=-\infty}^{\infty} S_{(n)}^{z(\pm)} e^{-in\Omega t}. \quad (11)$$

Employing this expansion in the dynamic equations (6) and (7), an infinite set of linear equations results in which the $S_{(n)}^{z(\pm)}$ are coupled with each other via the kernels. To obtain a solution for the spin variables, one has to terminate this infinite chain at a chosen order and then solve the resulting equations in a recursive way. However, in the limit of high frequencies, $\Omega \gg \Gamma$ (tunneling rate) and T (temperature), of interest in this letter, the ac-bias oscillates so fast that an electron experiences many cycles of the ac-bias during its presence inside the dot, and thus can not sense the details of the dynamics within one period \mathcal{T}_{ac} . In this non-adiabatic limit, one can approximately replace the kernels by a time-average with respect to the center-of-mass of time $\bar{\tau} = t + t'$:^{14,15}

$$C(R)(t, t') \approx C(R)^{(0)}(\tau) = \frac{1}{\mathcal{T}_{ac}} \int_0^{\mathcal{T}_{ac}} d\bar{\tau} C(R)(\tau, \bar{\tau}). \quad (12)$$

By the same token, one can retain only the stationary part of the spin variables and neglect the rapidly oscillatory parts, leading to detailed balance equations in a Markov approximation by making the replacement $\int_{-\infty}^t d\tau \Rightarrow \int_{-\infty}^{\infty} d\tau$ in Eqs. (6) and (7):

$$\begin{aligned} 0 = & -2 \left(C_{\omega}^{(0)}(-\Delta_0) + C_{\omega}^{(0)}(\Delta_0) \right) S_{(0)}^z \\ & + R_{\omega}^{(0)}(\Delta_0) - R_{\omega}^{(0)}(-\Delta_0), \end{aligned} \quad (13)$$

$$0 = \mp i\Delta_0 S_{(0)}^{\pm} - 2 \left[C_{\omega}^{(0)}(0) + C_{\omega}^{(0)}(\mp\Delta_0) \right] S_{(0)}^{\pm}. \quad (14)$$

Here, $C(R)_{\omega}^{(0)}(\omega)$ are the Fourier transforms of the time-averaged kernels $C(R)^{(0)}(\tau)$:

$$\begin{aligned} C_{\omega}^{(0)}(\omega) = & \pi g T \varphi\left(\frac{\omega}{T}\right) + \frac{\pi}{2} g T \sum_{n=-\infty}^{\infty} J_n^2\left(\frac{V_{ac}}{\Omega}\right) \\ & \times \left[\varphi\left(\frac{\omega + V + n\Omega}{T}\right) + \varphi\left(\frac{\omega - V - n\Omega}{T}\right) \right], \end{aligned} \quad (15)$$

$$R_{\omega}^{(0)}(\omega) = \pi g \left[1 + \sum_{n=-\infty}^{\infty} J_n^2\left(\frac{V_{ac}}{\Omega}\right) \right] \omega, \quad (16)$$

with $\varphi(x) \equiv x \coth(x/2)$ and $J_n(x)$ is the Bessel function of order n . To derive these equations (13)-(16), we use the relation

$$e^{ix \sin(\omega t)} = \sum_{n=-\infty}^{\infty} J_n(x) e^{in\omega t}. \quad (17)$$

The solution of Eq. (13) yields the nonequilibrium magnetization of the QD subject to an ac-bias voltage as

$$S_{(0)}^z = \frac{R_{\omega}^{(0)}(\Delta_0)}{2C_{\omega}^{(0)}(\Delta_0)}. \quad (18)$$

This formula is our central result, which can be regarded as a direct generalization of the dc nonequilibrium magnetization^{9,16,17} of a QD under a non-adiabatic high-frequency field. Obviously, it reduces exactly to previous results in absence of ac-bias, $V_{ac} = 0$.^{9,16,17} As an illustration, we exhibit in Fig. 1(a) the dependence of the magnetization, $S_{(0)}^z$, on dc bias-voltage for the driving frequency $\Omega/\Delta_0 = 0.5$. It should be noted that $S_{(0)}^z$ exhibits different behaviors with increasing ac-amplitude V_{ac} . For small dc bias voltage, the QD spin is fully polarized due to the nonzero external magnetic field, and it is gradually quenched with increasing dc bias voltage. Application of an ac bias tends to quench the spin polarization more rapidly. This tendency suggests that the ac bias plays a role in dephasing the electronic tunneling processes in a QD, as analyzed in Ref. 7.

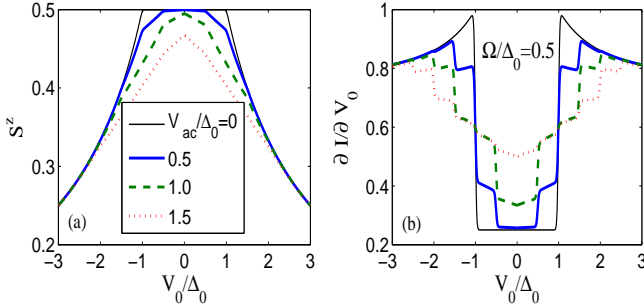


Figure 1: Nonequilibrium magnetization, $S_{(0)}^z$ (a); and the differential conductance, dI/dV_0 (in units of $2ge^2/h$) (b); as functions of dc bias-voltage, V_0 , for various amplitudes of the ac-bias with a fixed driving frequency $\Omega/\Delta_0 = 0.5$. The temperature we use in the calculation is $T/\Delta_0 = 0.01$.

Proceeding to the calculation of tunneling current, the current operator through the QD is defined as the time rate of change of charge density $N_{\eta} = \sum_{\mathbf{k}, \sigma} c_{\eta\mathbf{k}\sigma}^{\dagger} c_{\eta\mathbf{k}\sigma}$ in lead η : $I_{\eta}(t) = \dot{N}_{\eta}$. From linear-response theory we have

$$I(t) = \langle I_{\eta}(t) \rangle = -i \int_{-\infty}^t dt' \langle [J_{\eta}(t), H_I(t')]_- \rangle. \quad (19)$$

Because the dc component of current is easily measurable experimentally, we compute the time-averaged current $I = \frac{1}{T_{ac}} \int_0^{T_{ac}} dt I(t)$. Performing the same high-frequency and Markov approximations as above, we obtain the time-averaged currents:

$$I = 4\pi g \sum_{n=-\infty}^{\infty} J_n^2 \left(\frac{V_{ac}}{\Omega} \right) (V + n\Omega) + 2\pi g T S_{(0)}^z$$

$$\times \sum_{n=-\infty}^{\infty} J_n^2 \left(\frac{V_{ac}}{\Omega} \right) \left[\varphi \left(\frac{\Delta_0 - V - n\Omega}{T} \right) - \varphi \left(\frac{\Delta_0 + V + n\Omega}{T} \right) \right], \quad (20)$$

which is a generalization of Tien-Gordon-type formula in cotunneling current.¹⁴ It should be noted that our expression for photon-assisted cotunneling current is valid in the high frequency limit, whereas Flensberg derived a perturbative current under pump conditions in the low-frequency limit.⁸

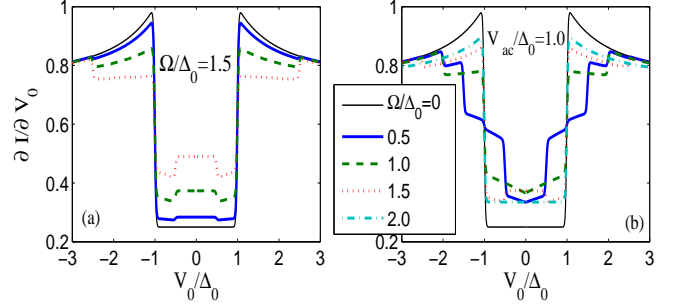


Figure 2: Calculated differential conductance vs. dc-bias. (a) results for a relatively high driving frequency $\Omega/\Delta_0 = 1.5$ for various ac-amplitudes, as in Fig. 1; (b) the ac-frequency dependence of dI/dV_0 for a fixed driving amplitude $V_{ac}/\Delta_0 = 1.0$. Other parameters are the same as in Fig. 1.

We plot the dc bias-voltage-dependent differential conductance, dI/dV_0 , in Figs. 1(b) and 2. Obviously, the differential conductance shows some satellites at $V_0 = \pm(\Delta_0 - n\Omega)$ superimposed on the characteristic jump at $V_0 = \pm\Delta_0$ in the presence of an ac-bias. These satellites arise physically from photon-assisted spin-flip cotunneling, i.e., albeit $|V_0| < \Delta_0$, the spin-flip cotunneling process can still become energetically activated by an electron absorbing photon quanta to compensate for the energy difference. Moreover, an overall suppression is observed with increasing ac-amplitude, which is qualitatively consistent with the experimental results.³ More interestingly, we find that dI/dV_0 exhibits a transition from peak-splitting to zero-bias-anomaly if the driving frequency is higher than Δ_0 . This behavior can be ascribed to the fact that a driving field with an appropriately high frequency can spur spin-flip cotunneling, notwithstanding $|V_0| < (\Omega - \Delta_0)$; in contrast, when the dc-bias increases to $\Delta_0 > |V_0| > (\Omega - \Delta_0)$, spin-flip events become inactive instead. The dI/dV_0 curve recovers peak-splitting behavior if $\Omega \geq 2\Delta_0$, as shown in Fig. 2(b).

In summary, we have generalized the generic Langevin equation approach to study photon-assisted cotunneling through an interacting QD in the non-adiabatic and high frequency regime, deriving explicit analytic expressions for the dc components of nonequilibrium magnetization and current with a generalized Tien-Gordon-type form. Our results show that applying an ac-bias is an important

method for tuning the I - V characteristics. Considering experiments² in which a static magnetic field $B = 11$ T is applied, $\Delta_0 \simeq 0.1$ meV, and the ac-frequency is $\Omega \sim 12 - 50$ GHz with ac-amplitude $V_{ac} \leq 0.15$ mV, all the parameters are easily accessible experimentally and they satisfy the non-adiabatic condition $\Omega \gg \Gamma$ (~ 30

μeV).

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